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THE DEVELOPMENT OF A 1990 GLOBAL INVENTORY FOR SO_x AND NO_x ON A $1^\circ \times 1^\circ$ LATITUDE-LONGITUDE GRID

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Abstract

Sulfur and nitrogen oxides emitted to the atmosphere have been linked to the acidification of water bodies and soils and perturbations in the earth's radiation balance. In order to model the global transport and transformation of SO_x and NO_x, detailed spatial and temporal emission inventories are required. Benkovitz *et al*. (1996) published the development of an inventory of 1985 global emissions of SO_x and NO_x from anthropogenic sources. The inventory was gridded to a 1°× 1° latitude-longitude grid and has served as input to several global modeling studies. There is now a need to provide modelers with an update of this inventory to a more recent year, with a split of the emissions into elevated and low level sources. This paper describes the development of a 1990 update of the SO_x and NO_x global inventories that also includes a breakdown of sources into 17 sector groups. The inventory development starts with a gridded global default EDGAR inventory (Olivier *et al*, 1996). In countries where more detailed national inventories are available, these are used to replace the emissions for those countries in the global default. The gridded emissions are distributed into two height levels (0-100m and >100m) based on the final plume heights that are estimated to be typical for the various sectors considered. The sources of data as well as some of the methodologies employed to compile and develop the 1990 global inventory for SO_x and NO_x are discussed. The results reported should be considered to be interim since the work is still in progress and additional data sets are expected to become available.

Introduction

Sulfur and nitrogen oxides emitted to the atmosphere undergo chemical reactions that lead to the formation and/or growth of aerosols that not only are responsible for acidification of water bodies and soils, but also play a role in the perturbation of the earth's radiation balance. In order to model the global transport and transformation of SO_x and NO_x, detailed spatial and temporal emission inventories are required. Data on anthropogenic emissions of SO_x and NO_x are generally available but are based on political units such as countries, states, or provinces. To be of use to the modelers, the emissions need to be allocated from the political units to a gridding system, such as a 1°× 1° latitude/longitude grid. Ideally, the location of all major industrial facilities would be known and their emissions assigned to the grid cell in which they are geographically located. Emissions from distributed sources, such as area and mobile sources, would then be allocated to a grid system based on a surrogate, which is representative of the emission source, the political unit and has the desired resolution of the gridding system.

A gridded 1985 global inventory of anthropogenic emissions of SO_x and NO_x has been conducted and documented by Benkovitz *et al.* (1996). The emission inventory presents annual emissions for a single vertical layer on a 1°× 1° latitude/longitude global grid. Benkovitz *et al.* (1996) estimate the total 1985 annual emissions of sulfur oxides to be 65000 kt S yr^{-1} and of nitrogen oxides to be 21000 kt N yr^{-1}. A complementary inventory, undertaken by Voldner *et al.* (1994), presents emissions of SO_x and NO_x by season using a two level vertical system (surface and above 100 m) with some resolution at the sector level. Both inventories were produced under the umbrella of the Global Emissions Inventory Activity (GEIA) of the International Global Atmospheric Chemistry (IGAC) Program.

The development of a global SO_x and NO_x inventory described in this paper is an update to 1990 of the prior 1985 GEIA inventory (Benkovitz *et al.*, 1996, and Voldner *et al.* 1994). One objective of this newer inventory is to resolve emissions by sectors at two heights (0-100 m and >100 m) to allow for greater flexibility for modelers and policy makers in determining the global effects of various control measures on different sectors. The grid system, as agreed upon by GEIA, is a 1°× 1° lat/long grid.

Sector Description

The sector divisions for the global 1990 SO_x and NO_x inventory are given in Table 1. Only SO_x and NO_x emissions from anthropogenic sources are considered in this study. The main source sectors are divided into power generation, fuel use and combustion, transportation, industrial processes, and land use. Each of these main emission sources is subdivided into one or more sector divisions for further refinement of the inventory. The type of source is identified in the last column. Elevated point sources are assumed to emit SO_x and NO_x emissions at a height ≥ 100 meters whereas all other source types are assumed to be surface releases.

Typically, emissions from major elevated point sources are assigned to a specific grid cell using the latitude/longitude coordinates of the facility. Emissions from low level point sources, area sources, and mobile sources typically require a surrogate for spatial allocation. In this study, the 1990 global population data set of Li (1996) is used as a default surrogate to geographically allocate the area source emissions. It should be noted, however, that not all sectors use population as a gridding surrogate if a more appropriate surrogate is available.

The population data set of Li (1996) is unique in that it allows for multiple countries as well as ocean to occupy a single grid cell. The data set contains lat/long grid cell numbers, the total population in each grid cell for each country sharing the grid cell, the ratio of the country's population in the grid cell to the total country's population, and country codes. A total of 210 countries are included in the data set. The 1990 global population data set, illustrated in Figure 1, is available for free download from the CGEIC web page (located at *www.ortech.ca/cgeic*).

Description Of Presently Available Data Sets

A listing of the available 1990 SO_x and NO_x emissions inventories is given in Table 2. The EDGAR data set is the only data set that has attempted to estimate global emissions of SO_x and NO_x. As such, it will be used as the global default data set. The EDGAR emissions are then replaced by more detailed, and presumably more accurate, regional or country SO_x and NO_x data where available. It should be noted that not all data sets have the same sector breakdown as that proposed for the current study. In such circumstances, the regional or country data is used only to replace the sectors that comprise the inventory while the EDGAR global default set is used to populate the missing sectors.

EDGAR Data Set

Olivier *et al*. (1996) have compiled a set of global "top-down" emission inventories of greenhouse gases and ozone depleting substance for anthropogenic sources on a per country basis and on a 1°×1° lat/long grid system. The purpose of the EDGAR database is to estimate the 1990 annual emissions per sector of direct and indirect greenhouse gases (CO_2, CH_4, N_2O, CO, NO_x, non-methane VOCs) and SO_x on a regional and grid basis.

The database is designed to be modular using a process approach whereby emissions are first calculated on a country basis by multiplying activity levels with emission factors for each compound which define the source strength. Regional emissions are generated by using the sources and regions defined by the processes and countries respectively. In addition, the EDGAR database uses a pre-defined spatial allocation function for each process to convert country emissions to a 1°×1° lat/long grid.

The EDGAR database consists of emissions resulting from fossil fuel related sources, biofuel combustion sources, industrial production and consumption processes, land-use related sources, and natural sources. Note that the EDGAR Version 2 database does not contain information on incineration, pulp and paper, and other industrial processes. These sector divisions, however, have been included in the current study to preserve as much detail from other regional inventories as possible.

Table 3 gives the total estimated mass of SO_x and NO_x emitted by each sector. Summing the sectors gives an annual 1990 emission of SO_x of 73540 kt S yr^{-1} and, for NO_x, an annual 1990 emission of 28450 kt N yr^{-1}. In comparing these figures with the 1985 emission inventory of Benkovitz *et al.* (1996), it must be kept in mind that the 1985 inventory did not address biomass burning. The EDGAR global totals, once biomass emissions are disregarded, represent approximately an 11% increase in elemental sulfur emissions and approximately a 17% increase in elemental nitrogen emissions over the 1985 inventory totals.

Figures 2 and 3 illustrate the geographical distribution of the total SO_x and NO_x emissions estimated by EDGAR respectively. To arrive at the plots, EDGAR country totals for emissions have been spatially allocated within each country using the surrogate population data set of Li (1996). Exceptions to this are air transportation, international shipping and biomass burning, which were originally generated on a 1°× 1° lat/long grid.

North America

The emission estimates for North America were generated using the Canadian Emissions Processing System (CEPS) (Scholtz *et al.*, 1999). CEPS, which is based on the urban-scale U.S. EPA's Emissions Processing System (EPS 2.0), has been extended to cover much larger geographical areas and multiple time zones. As input, CEPS uses the 1990 national inventory data of Canada and the United States. CEPS can produce gridded emissions for various domains, map projections and grid cell sizes resolved to time scales ranging from day-specific hourly to seasonal averages. Point, area, biogenic, and mobile source emissions are processed separately within CEPS to facilitate data tracking and to evaluate alternative control scenarios on predicted emissions.

For this project, the grid system used by CEPS is the 1°× 1° lat/long grid. In addition, Source Classification Codes (SCC) have been used to group emissions into the sectors described in Table 1.

CORINAIR 90 (Western Europe)

CORINAIR 90 is a study of emissions (SO_x, NO_x, non-methane VOC, CH_4, CO, CO_2, N_2O, and NH_3) for Western Europe (Grosslinger *et al*., 1996). The data set covers the twelve EU countries (Belgium, Denmark, France, the former West Germany, Greece, Ireland, Italy, Luxemburg, Netherlands, Portugal, Spain, and United Kingdom), the five EFTA-5 countries (Austria, Finland, Norway, Sweden, and Switzerland), the ten PHARE countries (Bulgaria, Czech Republic, Hungary, Poland, Romania, Slovak Republic, Estonia, Latvia, Lithuania, and Slovenia) as well as Croatia, Malta, and the former East Germany. The emission inventory is based on data supplied to the European Environment Agency from the individual participating countries. All country inventories were prepared according to a common format. CORINAIR 90 represents a complete, consistent, and transparent emission inventory for Western Europe.

Currently, only gridded total emissions on the 50 km × 50 km EMEP grid are available from the CORINAIR 90 study. It is planned to use sector totals by country and by 1°× 1° lat/long grid for the final 1990 global SO_x and NO_x inventory.

RAINS-ASIA (Asia)

RAINS-ASIA is an integrated program of assessment and policy analysis for the purpose of analyzing long-term strategies for acid rain problems at national and Asia-wide levels (Foell *et al.*, 1995). As part of the program, a preliminary SO_x and NO_x emission inventory has been developed for the base year of 1990 and covers the countries of East, South, and South-East Asia with particular focus on China, India, Thailand, and North and South Korea. The inventory subdivides emissions into industry, domestic, transport, and large and small power plants on a 1°× 1° lat/long grid system.

Former Soviet Union

Ryaboshapko *et al.* (1996) have complied an anthropogenic SO_x/NO_x emission inventory for the Former Soviet Union (FSU) for the years 1985 and 1990 on a 1°× 1° lat/long grid. Methodologies used in the inventory are a combination of “bottom-up” and “top-down” approaches.

Sulfur dioxide emissions from combustion of hard coal, brown coal, oil products and other fossil fuels as wells as from ferrous and non-ferrous metal production, sulfuric acid production and cement production have been estimated. A total of 721 point sources for SO_x have been considered although Ryaboshapko *et al*. (1996) consider point sources to be “separate enterprises, towns or cities which give substantial contribution to the total sulfur dioxide emission." For large cities, the total reported SO_x point source emission could thus be the sum of emissions from a number of single plants, boilers, power generating stations, etc.

For nitrogen oxide emissions, a separate inventory has been compiled for large power plants (242 point sources), small power plants, industrial boilers, residential combustion units, and for the transportation sector. In estimating the NO_x emissions from power generation stations, Ryaboshapko *et al.* (1996) have taken into account the high load demand on power stations in the FSU.

Australia

The Australian 1990 SO_x and NO_x inventory, given on a 1°× 1° lat/long grid, is divided into three broad categories: power generation point sources (31 in total), miscellaneous industrial point sources (41 in total), and diffuse or area sources which cover emissions from mobile and domestic sources (Carnovale, 1999).

Replacement Methodology

Each of the regional or country data sets that have been identified for use in replacing the EDGAR default data set have reported emissions aggregated to various sectors. One challenge of the current project is to map these regional data sets to the sector breakdown, as given in Table 1, using not only information in the regional data set, but also information in the EDGAR default data set as well as other sources of information. Two examples are presented next. In the first example, a simple substitution is shown for the emissions resulting from power generation (point source) for North America. In the second and more complicated example, the emissions from the Residential, Commercial, and Other (RCO) sector (area sources) in the former Soviet Union are examined.

Example 1: In order to generate gridded SO_x and NO_x emissions from the Canadian and US country totals reported in EDGAR, the population database of Li (1996) was used to spatially allocate the data. Figures 4 and 5 illustrate the end result for gridded SO_x and NO_x emissions respectively from power generation. Total emissions from power generation from North America, as estimated by EDGAR, are 7875 kt of elemental sulfur per year and 2050 kt of elemental nitrogen per year.

To generate the replacement data, the CEPS system was used. To isolate emissions resulting solely from power generation, a list of all SCC codes related to power generation were selected. In addition, since the locations of all power generation stations are part of the input data to the system, spatial allocation by a surrogate is not necessary. Figures 6 and 7 illustrate the resulting distribution of SO_x and NO_x emissions from power generation. The 1990 annual sum of power generation emissions for elemental sulfur and nitrogen is 7550 kt and 2130 kt respectively. This represents a difference for SO_x and NO_x emissions of approximately -4% and +4% over the EDGAR values respectively.

Although the total North American power generation emissions from EDGAR and CEPS are close, the patterns in Figures 4 and 5 are much different than those in Figures 6 and 7. By taking the EDGAR country totals and spatially allocating them by a population surrogate, the emissions are forced to follow the same distribution pattern as the population. The estimates from CEPS, given in Figures 6 and 7, clearly indicate that the location of large power plants do not necessarily correspond to large population centers. Thus, by using more refined regional inventories, the end result is a more accurate global emission inventory.

Example 2: The EDGAR default data set treats the Former Soviet Union (FSU) as one political entity. The FSU SO_x and NO_x emissions from the Residential, Commercial and Other (RCO) sector, as predicted by EDGAR, are given in Figures 8 and 9 respectively. The population data set of Li (1996) was used to spatially allocate the data by summing the entire population of all the FSU political subunits. Total emissions from the RCO sector for the FSU are at 1100 kt S yr^{-1} and 165 kt N yr^{-1}.

In the FSU data set of Ryaboshapko *et al.* (1996), the FSU is subdivided into its political subunits to account for some of the regional differences in the emission estimates. Data for the RCO sector is not explicitly given by Ryaboshapko *et al.* (1996). SO_x emissions are reported for point sources (Power Generation and Ferrous, Non-Ferrous, Cement and Chemical Processes) as well as for area sources (Industry, RCO, Road Transportation, and Non-Road Transportation). Using the EDGAR data set, a ratio of SO_x emissions from the RCO sector over the sum of SO_x emissions from the Industry, RCO, Road Transportation, and Non-Road Transport sectors was used to allocate a fraction of the FSU area source emissions to the RCO sector. NO_x emissions are, however, reported for small boilers, industrial sources and municipal economies which represents the sum of emissions from the Industry and RCO sectors as given in Table 1. To obtain an estimate for only the RCO sector, the EDGAR ratio of NO_x emissions from the Industry sector to the of NO_x emissions from the RCO sector was used to split the FSU NO_x emissions. The resulting emissions from the RCO sector for the FSU are given in Figures 10 and 11 for SO_x and NO_x respectively. The population data set of Li (1996) was again used to spatially allocate the emission data since the RCO sector is comprised of area sources and is likely well represented by population. The resulting patterns in Figures 10 and 11 are thus nearly identical to those found in Figures 8 and 9. The difference between the two inventories, however, is in the overall sum of emissions for the RCO sector. The FSU data set of Ryaboshapko *et al.* (1996) gives total estimated RCO emissions of 1320 kt S yr^{-1} and 195 kt N yr^{-1}. This represents a difference in emissions of +20% and +15% for SO_x and NO_x emissions in comparison to the EDGAR values.

When the sum of all sector emissions is compared between the two inventories, the SO_x emissions are surprisingly within one percent of each other (FSU data set = 11330 kt S yr^{-1}; EDGAR data set = 11443 kt S yr^{-1}). The NO_x emissions, however, are 32% larger in the FSU data set (4097 kt N yr^{-1}) than that in the EDGAR (3096 kt N yr^{-1}). Much of this difference can be attributed to the high load demand placed on FSU power generating stations as well as the transportation sector in which Ryaboshapko *et al.* (1996) have accounted for the poor quality of the FSU road systems. This, in turn, leads to lower average speeds and higher fuel consumption.

The 1985 inventory of Benkovitz *et al.* (1996) report total FSU SO_x and NO_x emissions at 15640 kt S yr^{-1} and 3170 kt N yr^{-1}. Comparing the 1990 data of Ryaboshapko *et al.* (1996) and the 1985 figures indicates an apparent 28% decrease in SO_x emission estimates and an apparent 30% increase in NO_x emissions over five years.

Conclusions

Although the final version of the 1990 global SO_x and NO_x inventory is not yet completed, it can be concluded that: maintaining sector detail, while more time consuming and onerous, will allow modelers and policy makers greater flexibility in using the data, and by incorporating regional or country emission inventories that attempt to account for the diversity and uniqueness within a coverage area, a more accurate global inventory results.

The final version of the 1990 global SO_x and NO_x inventory on a $1° \times 1°$ lat/long grid system is anticipated to be completed by January 2000. Once completed, the entire emission inventory will be available for download from the CGEIC web page (*www.ortech.ca/cgeic*)

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References

Benkovitz, C.M.; Scholtz, M.T.; Pacyna, J.; Tarrason, L.; Dignon, J.; Voldner, E.C.; Spiro, P.A.; Logan, J.A; Graedel, T.E. "Global gridded inventories of anthropogenic emissions of sulfur and nitrogen," *Journal of Geophysical Research*, 1996, *101(D22)*, 29,239-29,253.

Carnovale, F.,1999, Department of Primary Industries, Water and Environment, Hobart Tasmania, Australia. *Personal communication.*

Foell, W.; Amann, M.; Carmichael, G.; Chadwick, M.; Hettelingh, J.P.; Hordijk, L.; Dianwu, Z. *RAINS-ASIA: An Assessment Model for Air Pollution in Asia. Chapter 1*; World Bank, December 1995.

Grosslinger, E.; Radunsky, K.; Ritter, M. *CORINAIR 1990 Summary Report 1*; European Environment Agency, Copenhagen, Denmark, 1996.

Li, Y.F. *Global Population Distribution Database*; Report to the United Nations Environment Programme under UNEP sub-project FP/1205-95-12, March 1996.

Olivier, J.G.J.; Bouwman, A.F.; van der Maas, C.W.M.; Berdowski, J.J.M.; Veldt, C.; Bloos, J.P.J.; Visschedijk, A.J.H.; Zandveld, P.Y.J.; Haverlag, J.L., *Description of EDGAR Version 2.0: A set of global emission inventories of greenhouse gases and ozone-depleting substances for all anthropogenic and most natural sources on a per country basis and on 1°×1° grid,* National Institute of Public Health (RIVM), RIVM report #771060 002, Bilthoven, the Netherlands, 1996.

Ryaboshapko, A.G.; Brukhanov, P.A.; Gromov, S.A.; Proshina, Y.V.; Afinogenova, O.G., *Anthropogenic Emissions of Oxidized Sulfur and Nitrogen into the Atmosphere of the Former Soviet Union in 1985 and 1990*, International Meteorological School in Stockholm, 1996; Report CM-89.

Scholtz, M.T.; Taylor, A.; Ivanoff, A. "Preparation of 1990 North American emission inventory modelling files for AES regional air quality models"; Prepared for the Atmospheric Environment Service, Environment Canada, by Canadian ORTECH Environmental Inc., 1990

Voldner, E.C.; Li, Y.F.; Scholtz, M.T.; Davidson, K.A. "1°×1° Global SO_x and NO_x 2-Level Inventory Resolved Seasonally Into Emission Sectors and Point and Area Emission Sources," Presented at the *Fifth International GEIA Workshop on Global Emission Inventories*, Fuji-Yoshida, Japan, September 1994.

Table 1. Sector division for the global 1990 SO_2/NO_x inventory.

Main Source	Sector Division	Source Type
Power generation	Power generation	Elevated point source
Fuel use and combustion	Industry (including other transformation sectors such as refineries, coke ovens, gas works, etc.)	Low level point source
	Residential, commercial and other (RCO)	Area
	Incineration	Area
Transportation	Road	Mobile
	Non-road (*i.e.* rail)	Mobile
	Air (below one kilometer)	Mobile
	International shipping	Mobile
Industrial processes	Iron and steel (excludes coke ovens and blast furnaces; for NO_x, sum of sinter and crude steel production; for SO_x, sum of sinter production)	Elevated point source
	Non-ferro (including primary and secondary copper, lead, zinc, and aluminum)	Elevated point source
	Chemicals (for NO_2, sum of nitric acid and ammonia production; for SO_2, sum of H_2SO_4 production)	Low level point source
	Cement	Elevated point source
	Pulp and paper	Elevated point source
	Other	Low level point source
Landuse	Biomass burning	Area

Table 2. Summary of available emission inventory data sets.

Data Set	Coverage Area	Grid System
EDGAR Version 2	Global Default	1°×1° lat/long
CEPS 1990	Canada and U.S.A.	1°×1° lat/long
CORNAIR 1990	Western Europe	50 km × 50 km EMEP grid
Soviet Union	Former Soviet Union	1°×1° lat/long
RAINS-ASIA	Asia	1°×1° lat/long
Australia	Australia	1°×1° lat/long

Table 3. EDGAR global SO_x and NO_x emissions by sector.

Main Source	Sector Division	SO_x (kt S yr^{-1})	NO_x (kt N yr^{-1})
Power generation	Power generation	27924.0	6279.1
Fuel use and combustion	Industry (including other transformation sectors such as refineries, coke ovens, gas works, etc.)	20737.0	4318.9
	Residential, commercial and other (RCO)	6939.5	2109.8
	Incineration	n.a.	n.a.
Transportation	Road	2000.5	9581.8
	Non-road (*i.e.* rail)	462.5	341.5
	Air (0 to 1 km only)	9.6	182.4
	International shipping	2465.0	228.9
Industrial processes	Ferro (Iron and steel excluding coke ovens and blast furnaces)	434.0	291.5
	Non-ferro (primary and secondary copper, lead, zinc, & Al)	8606.5	
	Chemicals	1749.5	131.6
	Cement	914.5	1042.7
	Pulp and paper	n.a.	n.a.
	Other	n.a.	n.a.
Landuse	Biomass burning	1296.5	3944.1
Total		73539.1	28452.5

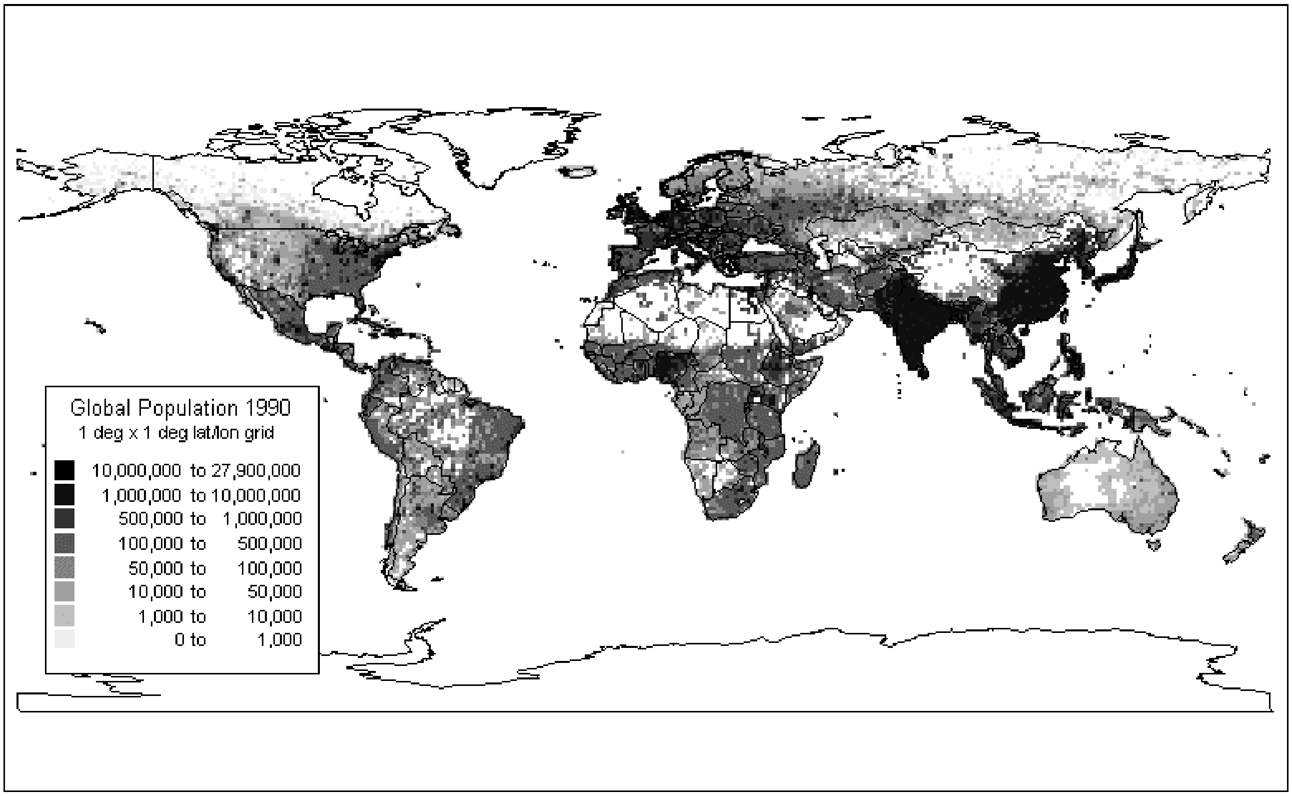

Figure 1. Global 1990 population data set of Li (1996).

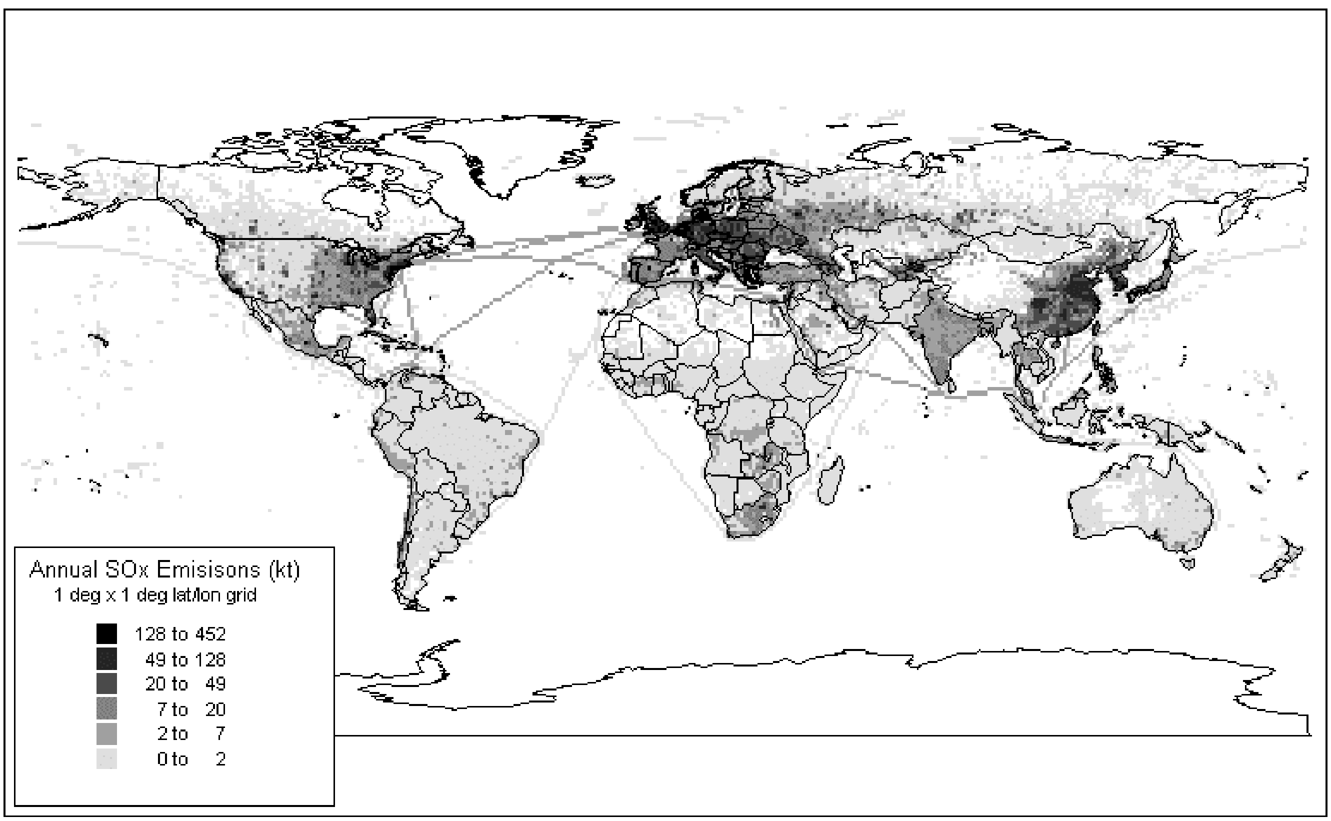

Figure 2. Default global SO_x emissions from EDGAR with all sectors included.

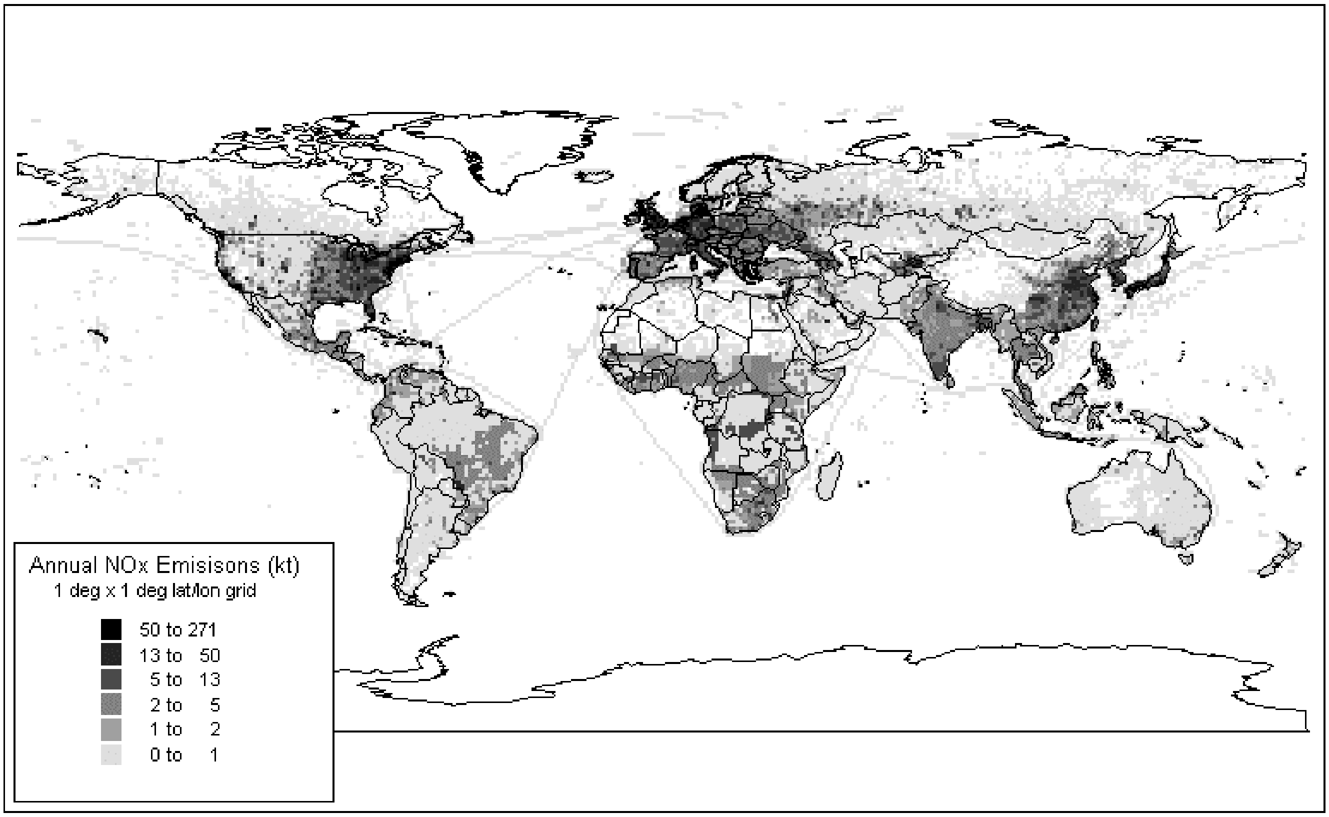

Figure 3. Default global NO_x emissions from EDGAR with all sectors included.

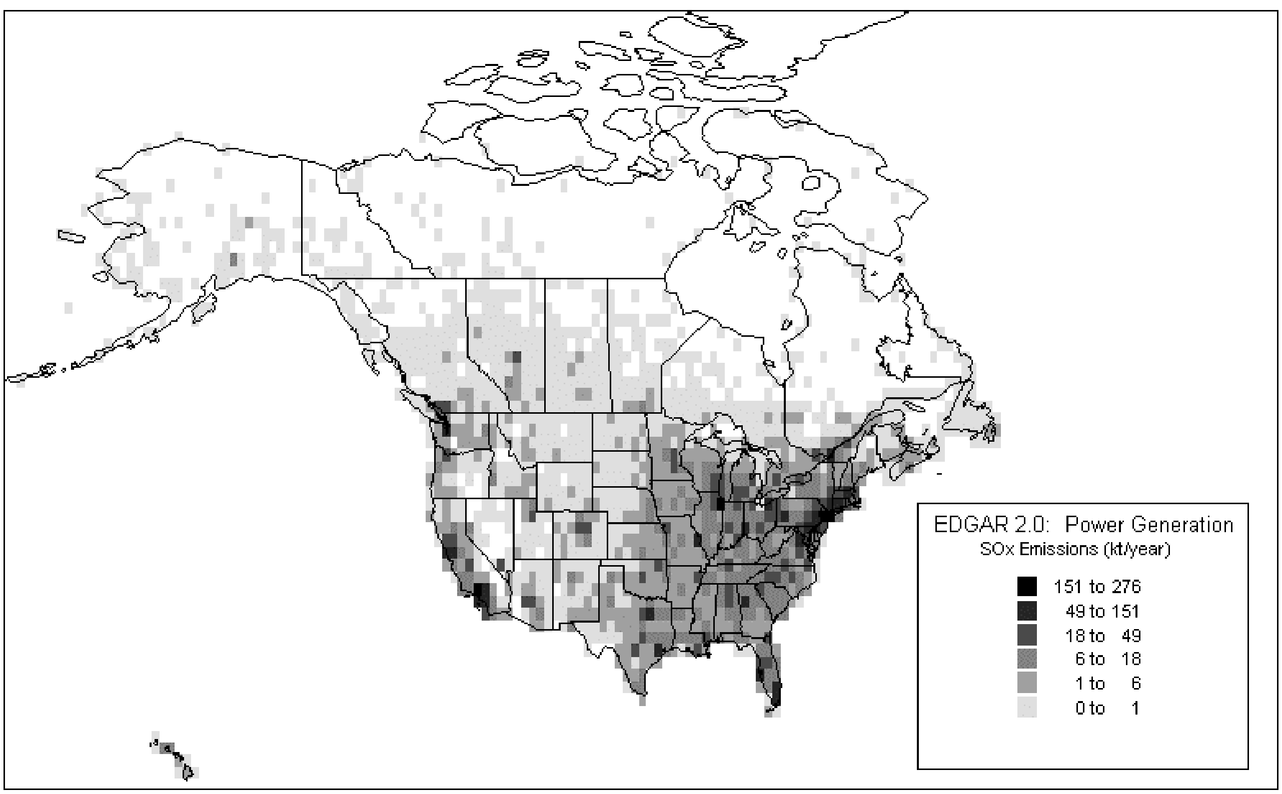

Figure 4. SO_x emissions from power generation in North America: EDGAR data set.

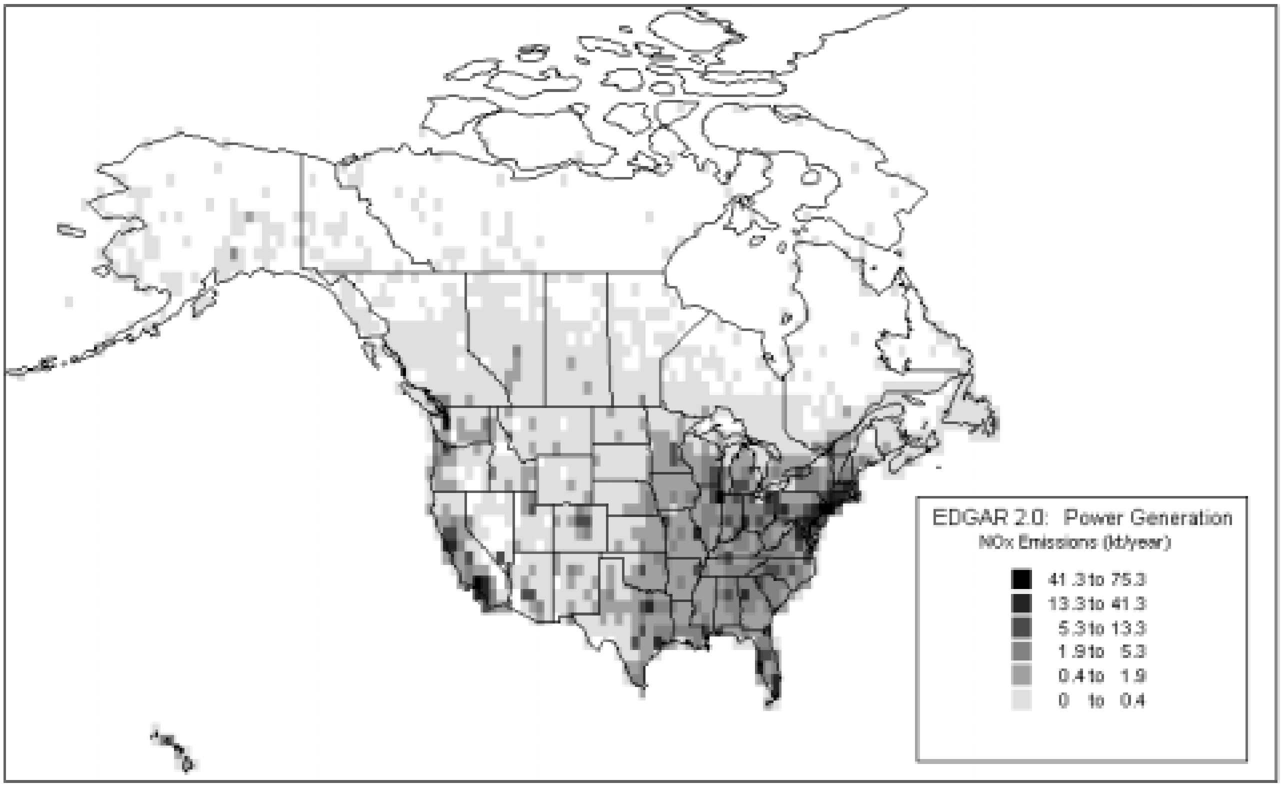

Figure 5. NO_x emissions from power generation in North America: EDGAR data set.

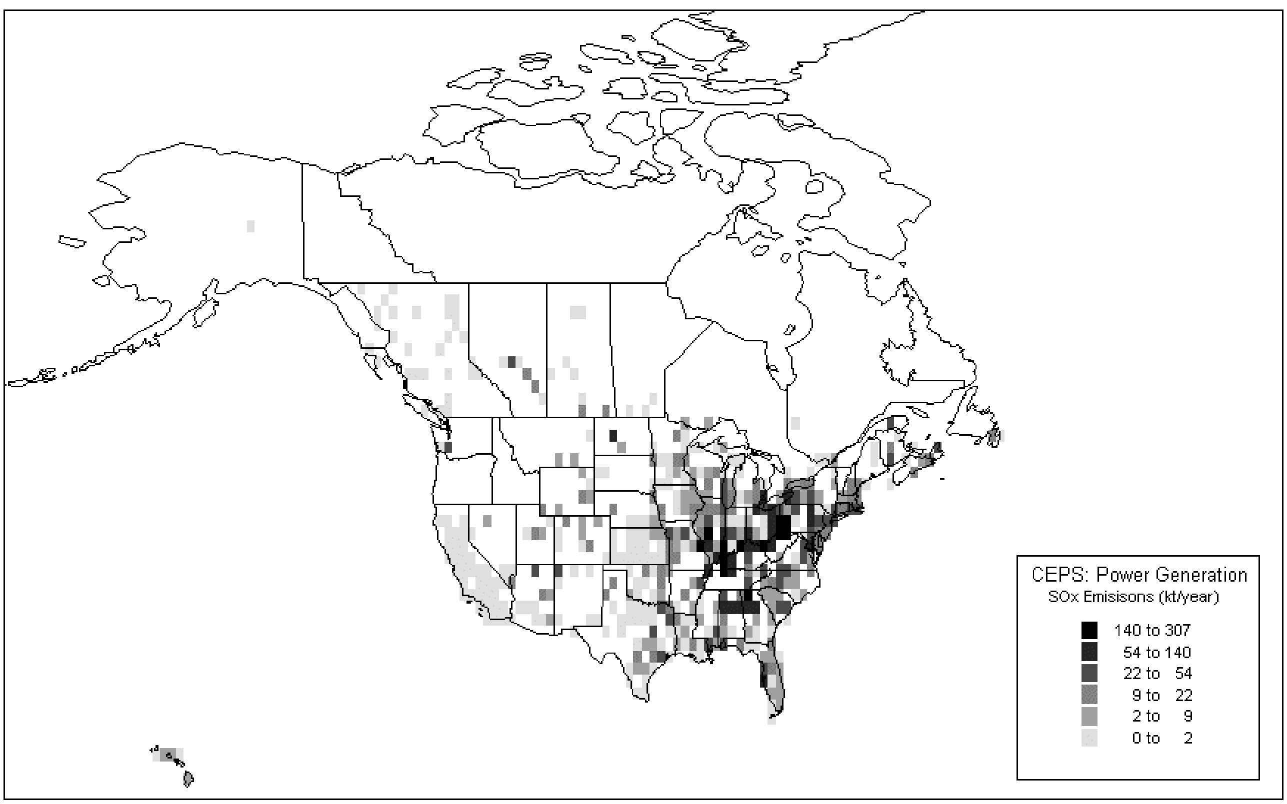

Figure 6. SO_x emissions from power generation in North America: CEPS data set.

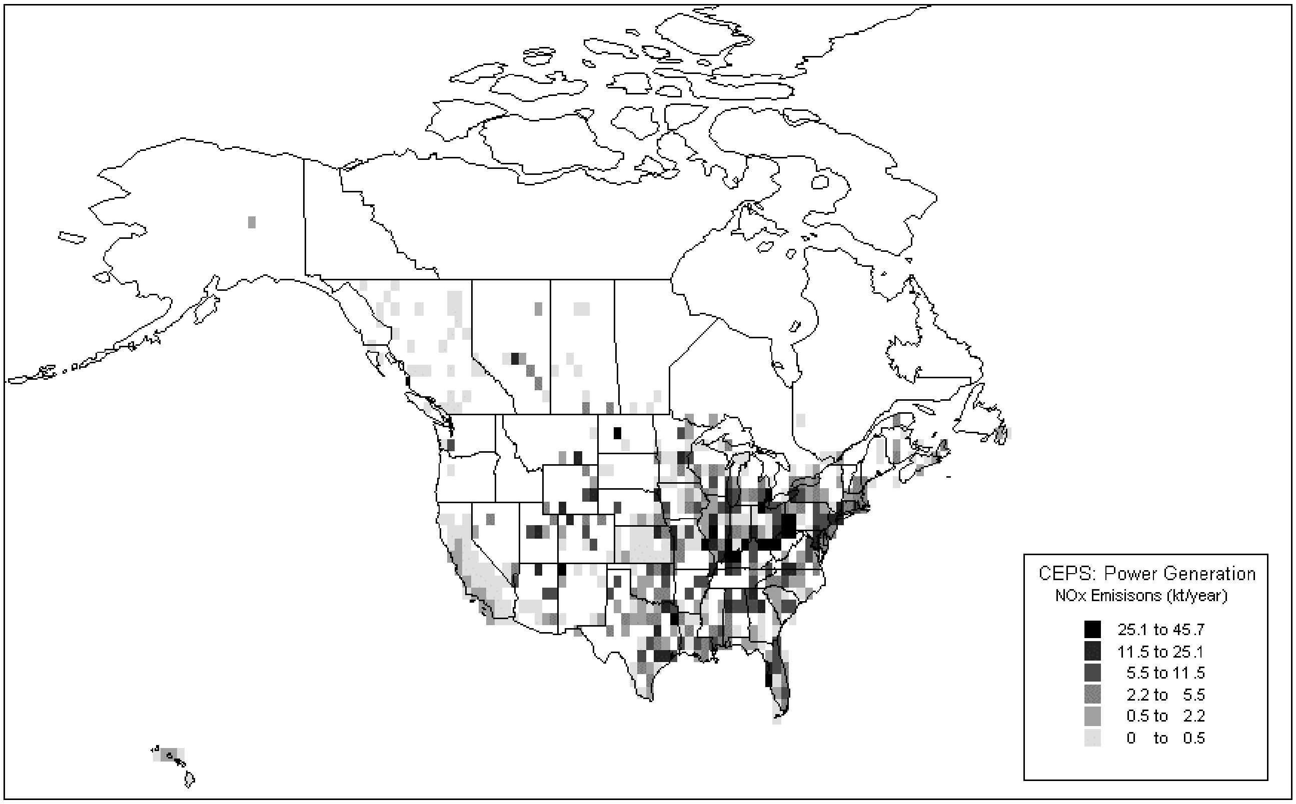

Figure 7. NO_x emissions from power generation in North America: CEPS data set.

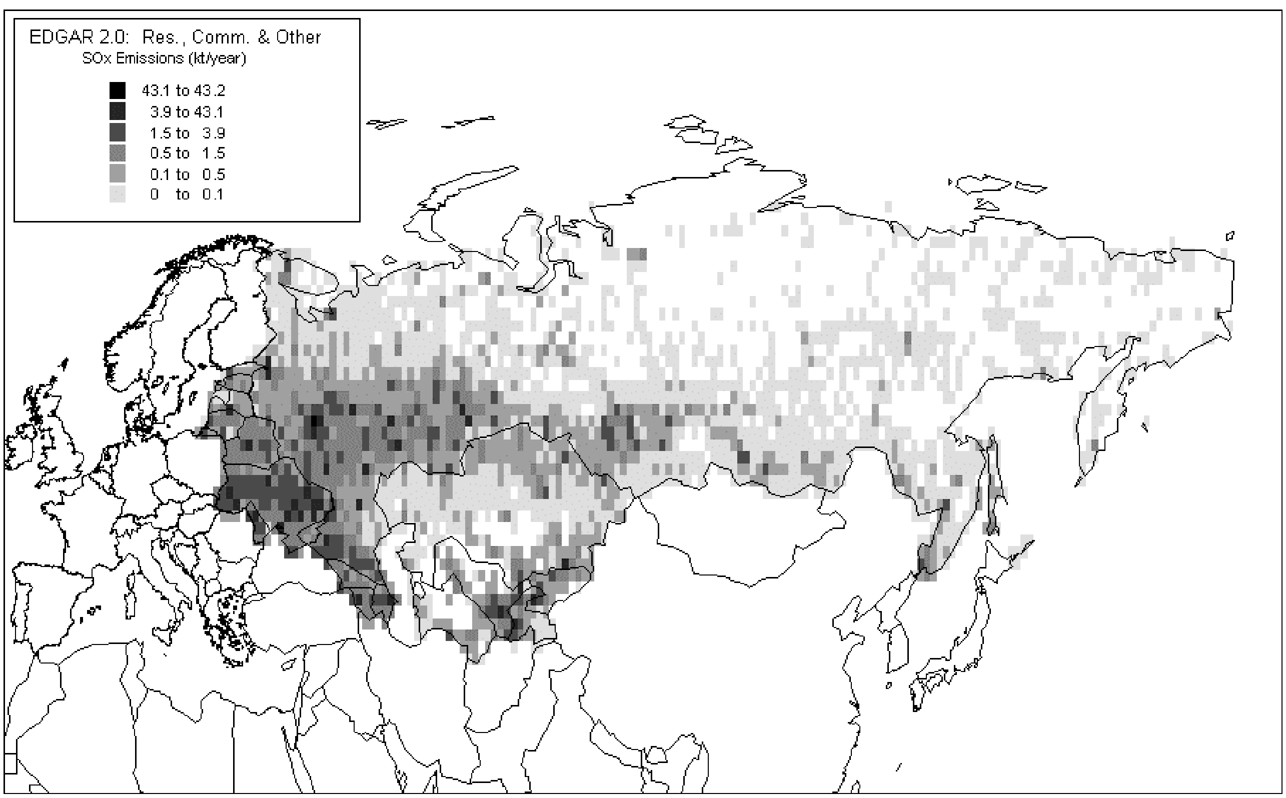

Figure 8. SO_x emissions from Residential, Commercial and Other (RCO): EDGAR data set.

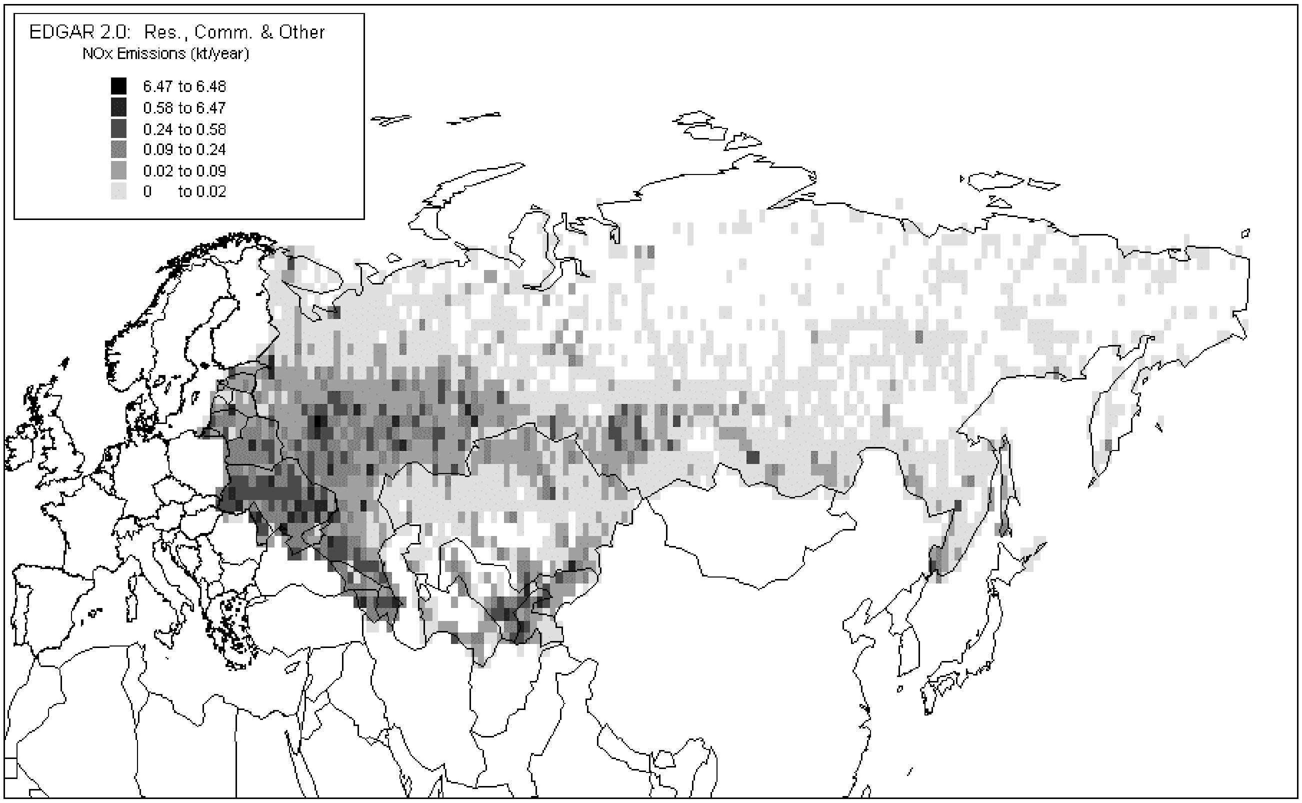

Figure 9. NO_x emissions from Residential, Commercial and Other (RCO): EDGAR data set.

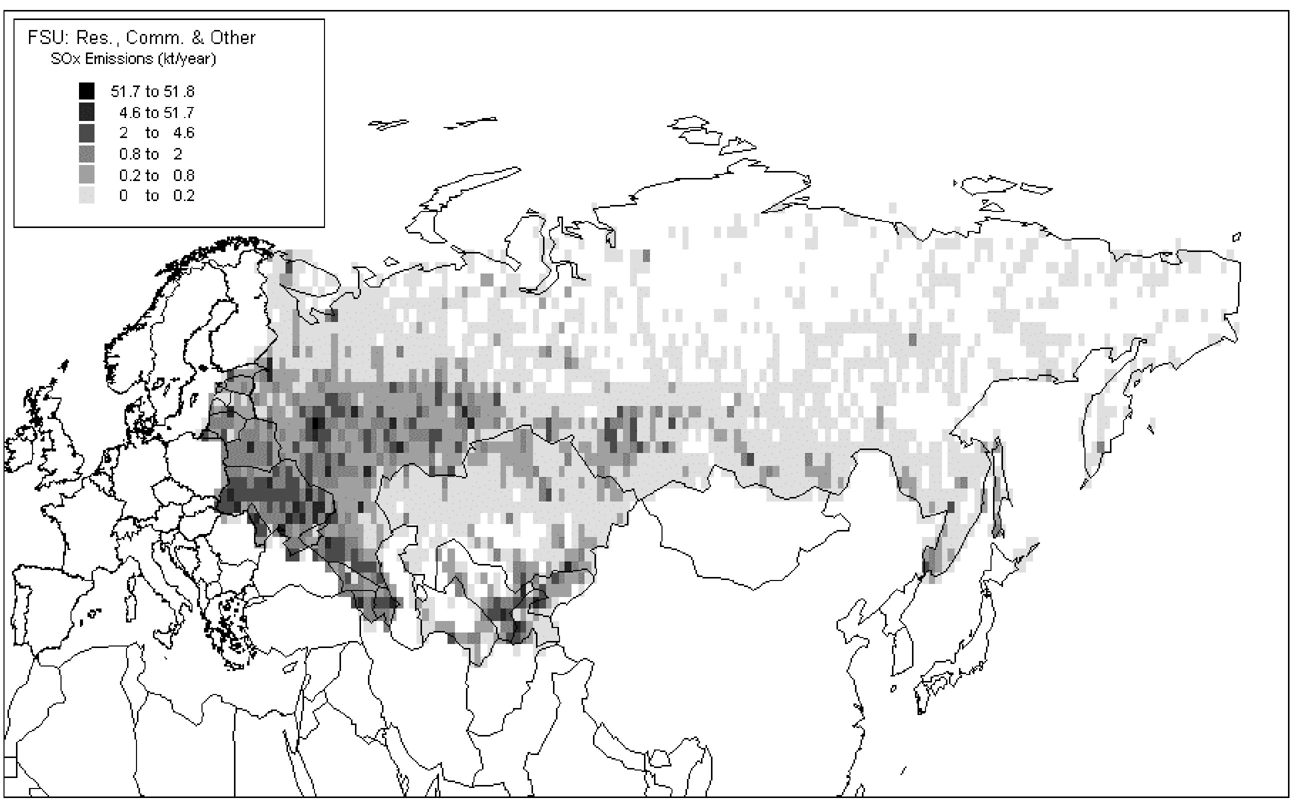

Figure 10. SO_x emissions from Residential, Commercial and Other (RCO): FSU data set.

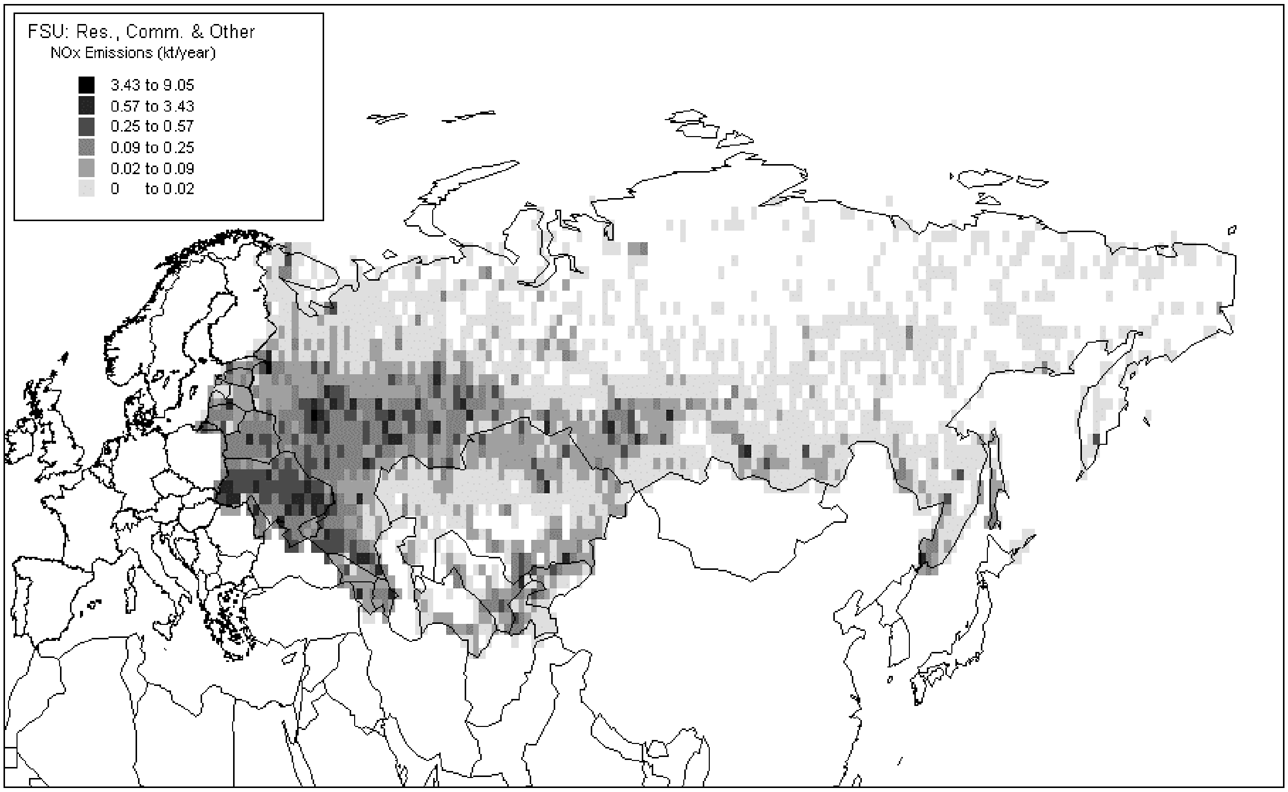

Figure 11. NO_x emissions from Residential, Commercial and Other (RCO): FSU data set.